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SPALLATION REACTIONS OF CALIFORNIUM-252 WITH HELIUM IONS

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### Authors

Sikkeland, Torbjorn  
Amiel, Saadia  
Thompson, Stanley G.

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Torbjörn Sikkeland,\* Saadia Amiel,\*\* and Stanley G. Thompson

Printed for the U. S. Atomic Energy Commission

\* On leave from JENER, Norway.

\*\* On leave from Israel Atomic Energy Commission, Hakirya, Tel Aviv, Israel.

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ABSTRACT

The excitation functions for the  $(\alpha, n)$ ,  $(\alpha, 2n)$ ,  $(\alpha, 3n)$ ,  $(\alpha, 4n)$ ,  $(\alpha, pn)$ , and  $(\alpha, p2n)$  reactions of the isotope of  $\text{Cf}^{252}$  with 20- to 40-Mev helium ions have been measured. A brief discussion of the reactions studied is given.

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## INTRODUCTION

Previous publications from this laboratory have given results for excitation functions of spallation reactions on heavy elements in the region of  $Z > 90$  with helium ions.<sup>1-5</sup> The heaviest element studied in detail was Cf<sup>249</sup>.<sup>6</sup> The purpose of the work presented here was to obtain information concerning the reactions of helium ions with the heaviest nucleus available for such studies, and to observe any influence of the 152-neutron subshell on the yield of the  $(\alpha, 4n)$  reaction product Fm<sup>252</sup>. An additional objective of the work was to verify previous values for some decay properties of the isotopes produced.

## EXPERIMENTAL

The Cf<sup>252</sup> was prepared from Pu<sup>239</sup> by successive neutron-capture and beta-decay reactions in the Material Testing Reactor. The separated californium containing Cf<sup>249</sup>, 250, 251, and 252 was put back into the reactor and irradiated again with neutrons for approximately 8 months to convert all light californium isotopes into Cf<sup>252</sup>. The Cf<sup>252</sup> used for the target contained  $< 2\%$  of Cf<sup>254</sup> and negligible amounts of the other isotopes.

The Cf<sup>252</sup> ( $2.4 \times 10^{-3}$  microgram) was chemically purified and electroplated as a uniform thin deposit on a 0.002-inch-thick gold foil in an area of  $0.05 \text{ cm}^2$ . A measured amount of Cm<sup>244</sup> was added in the electroplating step in order to use its well-known  $(\alpha, 2n)$  excitation function as an internal standard for the cross-section measurements. The bombardments were carried out at the 60-inch cyclotron of the Crocker Laboratory, using a catcher-foil technique with the deflector-channel target assembly previously described.<sup>5</sup>

The energies of the incident helium ions were reduced to the desired values with aluminum absorbers. The recoil products were collected on gold foils of about  $5 \text{ mg/cm}^2$  thickness. The gold was dissolved in a mixture of hydrochloric and nitric acids. The gold was then separated from the spallation products by sorption on a column of Dowex A-1 anion-exchange resin and the spallation products were electroplated on a platinum disc. The fermium, einsteinium, and californium isotopes were identified by measurements of their decay properties<sup>2,6,7</sup> in an ionization-grid chamber with a 48-channel alpha-particle pulse-height analyzer. The decay of the various alpha-particle peaks was usually followed through several half lives. In some cases the fermium, einsteinium, and californium were separated from each other in the final step by elution with ammonium-alpha-hydroxy isobutyrate from a column of Dowex 50 cation-exchange resin.<sup>8</sup>

#### RESULTS

The cross sections measured are shown in Figs. 1 and 2, as a function of the energy of the bombarding helium ions. The upper limit for the cross section of the  $(\alpha, p)$  reaction was found to be 0.5 millibarn. The cross section for the production of  $\text{E}^{254}$  (long-lived isomer) was always less than 6 millibarns.

The isotope  $\text{Fm}^{253}$  was found (by following the decay of its alpha particles) to have a half life of  $3.0 \pm 0.2$  days. Its most abundant alpha-particle group had an energy of  $6.95 \pm .05$  Mev. A lower alpha group of about 6.90 Mev energy seemed to be present, in which case the measured ratio of the 6.95 to the 6.90 peak would be of the order of 4. The  $\text{Fm}^{253}$  decay was also followed by observing the corresponding growth of its electron-capture daughter,  $\text{E}^{253}$ , in the fermium fraction. The rate of formation of  $\text{E}^{253}$  was consistent with the over-all half life of  $3 \pm 0.2$  days for  $\text{Fm}^{253}$ , and the amount of it formed gives an electron-capture to alpha-decay branching ratio of  $8.5 \pm 1.0$ . A previous publication on  $\text{Fm}^{253}$  reported an alpha-particle energy of  $6.94 \pm$  Mev, a branching ratio of 8.5, and a half life of  $4.5 \pm 1.0$  days.<sup>7</sup> Confirmation of the  $\text{Fm}^{253}$  mass assignment was obtained in the excitation function for its production. This excitation function had the expected shape for an  $(\alpha, 3n)$  reaction in the heavy region. The other isotopes detected in this work were found to have decay properties in agreement with previous work.<sup>2,6,7</sup>

## DISCUSSION

The broad features of the excitation functions shown in Figs. 1 and 2 are similar to those of other very heavy isotopes, the interpretation of which has been discussed by other authors.<sup>1-5</sup> The statistical errors are indicated for the points shown. The measured values for the  $(\alpha, 2n)$  cross sections lie on a smooth curve. The errors in the determination of this excitation function should be small because the recoil collection efficiencies for the  $(\alpha, 2n)$  reaction product in the reference standard should be the same.

The products of the  $(\alpha, n)$  and  $(\alpha, 4n)$  reactions, namely  $\text{Fm}^{255}$  and  $\text{Fm}^{252}$ , have similar half lives and alpha-decay energies. Thus satisfactory resolution of these isotopes could not be accomplished in the pulse-height analyzer used, and the reaction yields are rather uncertain. The curve shown in Fig. 1 for the  $(\alpha, n)$  reaction, therefore, includes the contribution from the  $(\alpha, 4n)$  reaction. However, the  $(\alpha, n)$  curve shown must give the true values for the  $(\alpha, n)$  cross sections below the energy corresponding to the threshold for the  $(\alpha, 4n)$  reaction, which is 31 Mev. An extrapolation of the  $(\alpha, n)$  curve above 31 Mev was made, based on the shape of the  $(\alpha, n)$  excitation functions of  $\text{U}^{235}$  obtained by Vandenbosch.<sup>9</sup> Subtraction of the  $(\alpha, n)$  cross sections obtained in this way from the sum of the measured  $(\alpha, n)$  and  $(\alpha, 4n)$  yield gives an  $(\alpha, 4n)$  cross section at 46 Mev of about  $1.2 \pm 1$  millibarns. The peak of the  $(\alpha, 4n)$  excitation function should occur at about 43 Mev, and it would be of interest to extend the measurements to higher energies in order to obtain more precise values for the cross sections and thus more information about the influence of the 152-neutron subshell.

The cross section for the  $(\alpha, 3n)$  reaction products at the maximum bombarding energy appears to be about 100% higher than the value obtained by an extrapolation of the smooth curve. This may be explained as probably due to a higher recoil-collection efficiency for this product relative to the  $(\alpha, 2n)$  product resulting from a change in thickness of the target. The excitation function for the  $(\alpha, 3n)$  reaction may be interpreted in a manner that gives some information on the nuclear temperature and the competition between neutron emission and fission in the intermediate steps of the reaction process. This interpretation depends on the assumption that direct interaction processes may



be neglected. Then we may express the cross section thus:

$$(a) \quad \sigma(\alpha, 3n) = \sigma_c (\bar{G}_{3n})^3 P_{3n},$$

where  $\sigma(\alpha, 3n)$  is the cross section for the  $(\alpha, 3n)$  reaction, and  $\sigma_c$  is the total cross section for compound-nucleus formation as taken from Reference 10, assuming  $r_0 = 1.5 \times 10^{-13} A^{1/3}$  cm. This assumption has given the best fit for other helium-ion-induced reactions in heavy elements.<sup>1</sup>

$$(b) \quad \bar{G}_{3n} = (G_1 \cdot G_2 \cdot G_3)^{1/3},$$

$$\text{where } G_i = \left( \frac{\Gamma_n}{\Gamma_{t i}} \right) \approx \left( \frac{\Gamma_n}{\Gamma_n + \Gamma_f} \right)_i,$$

$\Gamma_n$  is the level width for neutron emission, and  $\Gamma_f$  is the width for fission.

$$(c) \quad P_{3n} = I(\Delta_3, 3) - I(\Delta_{3f}, 5),$$

according to Jackson,<sup>11</sup> and is the probability for boiling out exactly three neutrons;  $I(z, n)$  is Pearson's incomplete gamma function.

$$(d) \quad \Delta_3 = \frac{E^* - \sum_3 B_i}{T},$$

where  $E^*$  is the excitation energy of the compound nucleus,  $\sum_3 B_i$  is the sum of the neutron binding energies taken from Glass et al.,<sup>12</sup>  $T$  is the nuclear temperature assumed to be constant through the evaporation process.

$$(e) \quad \Delta_{3f} = \frac{E^* - \sum_3 B_i - E_{th}}{T},$$

where  $E_{th}$  is the activation energy for fission for the residual nucleus. This is taken to be 4.2 Mev from Reference 9.

Assuming  $T$  and  $\bar{G}_{3n}$  to be independent of excitation energy, the best fit to the measured  $(\alpha, 3n)$  excitation function is obtained for a nuclear temperature of 1.9 Mev and  $\bar{G}_{3n} = 0.14 \pm .02$ .

The points for the  $(\alpha, p2n)$  excitation function lie on a smooth curve, suggesting that no large changes in the recoil-collection efficiency relative to the  $(\alpha, 2n)$  collection efficiency occurred through the series of experiments done. The  $(\alpha, p2n)$  reaction has been demonstrated to occur mainly through an  $(\alpha, t)$  stripping mechanism<sup>3</sup> and the corresponding excitation function studied in the present work is consistent with the same mechanism.

The peculiar shape of the excitation-function curve for the  $(\alpha, pn)$  reaction might be explained in part by variation in the ratios of the two isomers as the bombarding energy changes. Therefore it is difficult to decide whether these results are inconsistent with the results of other work.<sup>13</sup>

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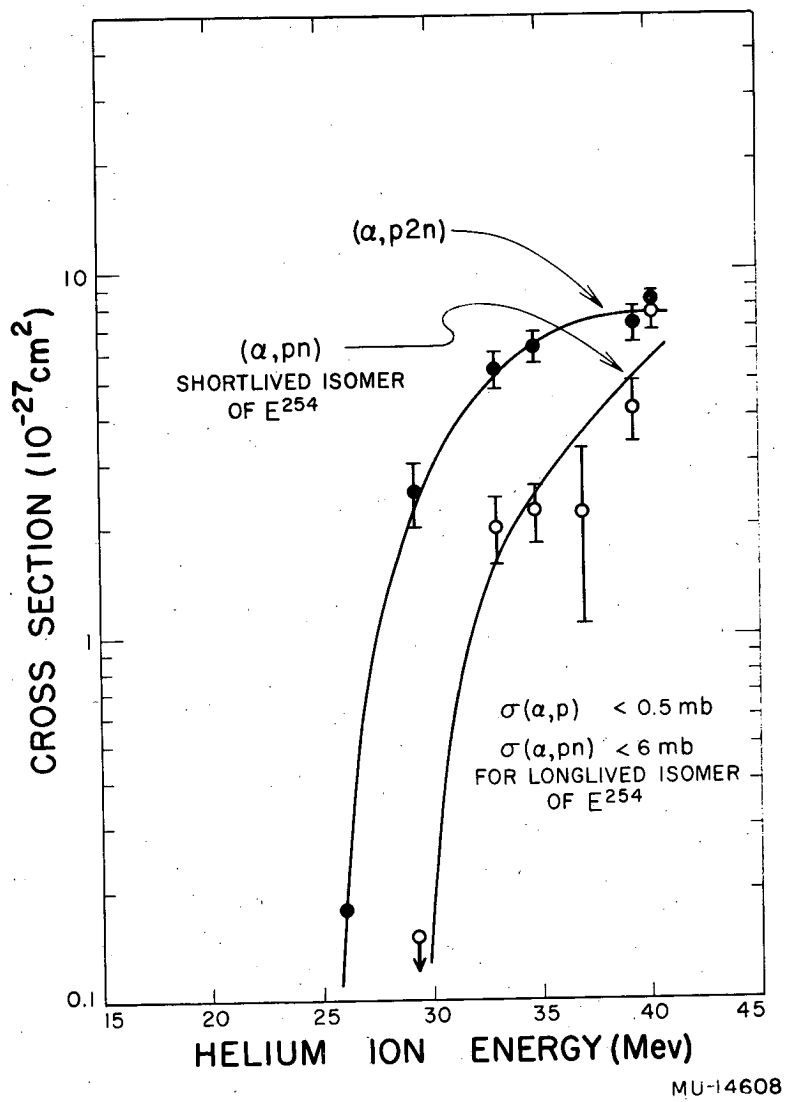


Fig. 1. Excitation functions for  $\text{Cf}^{252} (\alpha, pxn)$  reactions

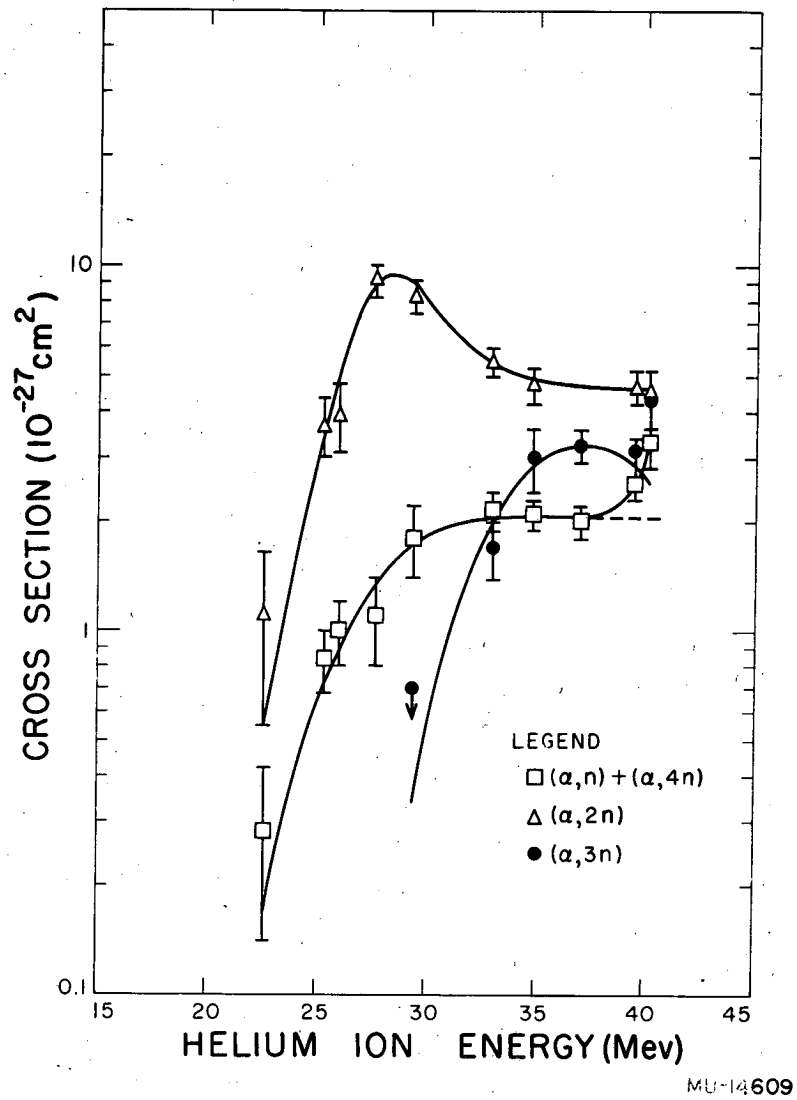


Fig. 2. Excitation functions for  $\text{Cf}^{252}(\alpha, xn)$  reactions